

Terahertz Electrodynamics of 180° Domain Walls in Thin Ferroelectric Films

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We investigate oscillation dynamics of a periodic structure of the 180° domain walls in nanometrically thin substrate-deposited ferroelectric films and superlattices. We calculate dynamic permittivity of such structures and reveal a collective resonance mode, which in the typical ferroelectric compounds, $\text{PbTiO}_3/\text{SrTiO}_3$, lies in the sub- and low THz frequency range of $0.3 \div 3$ THz. We propose the reflection-absorption spectroscopy experiments to observe this mode.

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A high-frequency dynamic response of ferroelectric materials is widely used in advanced electronic and optoelectronic applications. A wide range of operational frequencies that spans several decades from few kilohertz to tens terahertz ($1 \text{ THz} = 10^{12} \text{ Hz}$) is provided by the multiscale organization of spontaneous polarization. Whereas the ac response properties in radio- and microwave diapasons $10^4 \text{ Hz} \div 0.3 \text{ THz}$ are mostly due to the relaxation dynamics of the domain walls (DWs) and polar clusters, the far-infrared spectral region of $3 \div 30 \text{ THz}$ is governed by the soft-mode vibrations of the polar ions. The frequency window of $0.3 \div 3 \text{ THz}$ is less explored, hence the quest posed by the needs of the emergent THz technologies in the compact and tunable devices working in this THz frequency range [1, 2].

In this Letter we demonstrate that oscillations of the periodic 180° polarization domains in ferroelectric thin films exhibit resonance behavior in the exactly this $0.3 \div 3 \text{ THz}$ frequency range. Thus, ferroelectric films containing polarization domains are a fertile ground for designing sub- and low THz radiation range devices.

The regular polarization domain structures, that form in order to cancel a macroscopic depolarizing field, which would have possessed large energy proportional to the volume of the system, were first predicted in earlier works by Landau [3, 4] and Kittel [5] in the context of ferromagnetic systems. However their existence in the ferroelectrics have long been considered as barely possible until recent direct experimental evidences for equilibrium 180° stripe domains in strained ferroelectric thin films of PbTiO_3 (PTO) deposited on the SrTiO_3 (STO) substrate [6, 7] and in PTO/STO superlattices [8, 9] became available. The newly discovered domain structures exhibited behaviors in a good accord with the theoretical predictions [10–15] and were considered to be extremely suitable for the future nanodomain-based electronics [16].

Although the low-frequency dissipative motion of DWs in macroscopic ferroelectric samples has been receiving a theoretical attention [17], the crossover from the relaxation dynamics to the resonance was revealed only recently in the very thin ($2 \div 7 \text{ nm}$) films of $\text{PbTi}_{0.6}\text{Zr}_{0.4}\text{O}_3$ (PZT) by *ab-initio* simulations [18] at sub-THz frequencies of $0.3 \div 1 \text{ THz}$. In what follows we develop the theory of collective vibrations of periodic 180° domain structure and calculate its dynamical permittivity. This allows us to generalize the results of [18] for the arbitrary ferroelectric films to optimize the oscillation parameters for the best application conditions. In particular, (see Fig. 1b), we demonstrate that the domain resonance in traditional PTO/STO systems can be achieved at higher frequencies, with smaller relative damping and for wider range of film thickness. We propose that these oscillations can be detected by methods of reflection-absorption spectroscopy [19].

A graphic illustration of the response mechanism of a domain structure to the applied ac electric field is presented in Fig. 1a. A ferroelectric film of the thickness $2a_f$ is sandwiched between two paraelectric layers of the thickness a_p and placed into the biased capacitor. Then a periodic structure of the domain stripes aligned along the y -axis forms in the ferroelectric layer. The polarization axis z is perpendicular to the film plane, while the in-plane DW motion occurs along the x -axis [20]. Note that the described geometry is formally equivalent to the repeating system of paraelectric and ferroelectric layers of thickness of $2a_f$ and $2a_p$ [12]. Therefore, our consideration is equally applicable to the corresponding ferroelectric/paraelectric superlattices.

The dielectric properties of the system are described by the intrinsic permittivities of the ferroelectric film along and across the polar axis, ε_{\parallel} , ε_{\perp} , and by the permittivity of the paraelectric layers, ε_p , that are assumed to be almost frequency independent in the interesting for us sub- and low-THz range. In the absence of the field, the up- and down- oriented spontaneous polarizations of alternating domains, $\pm P_s$, compensate each other. The equilibrium domain width, d is given by the famous Landau-Kittel square root law [3–5], presented in the universal form [13],

$$d \simeq \left(\frac{\varepsilon_{\perp}}{\varepsilon_{\parallel}} \right)^{1/4} \sqrt{3.53 \zeta \delta (2a_f)} \quad (1)$$

and exemplified for PTO films in Fig. 1b. Here the DW thickness, δ , is about 1 nm [21], $\zeta = 1 + \varepsilon_p / (\varepsilon_{\parallel} \varepsilon_{\perp})^{1/2}$ and the values of intrinsic permittivities along and across the polarization direction, ε_{\parallel} , ε_{\perp} and of paraelectric layer, ε_p , are specified below; in the experimental range $1 \leq \zeta \leq 4$.

The applied field induces the net polarization of the ferroelectric layer, $\bar{\mathbf{P}} \parallel \mathbf{z}$, that can be conveniently decomposed into two contributions, $\bar{\mathbf{P}} = \bar{\mathbf{P}}_i + \bar{\mathbf{P}}_{dw}$. The intrinsic contribution, $\bar{\mathbf{P}}_i$ is provided by increasing of the up- and decreasing of the down-oriented polarization inside the alternating domains with the conservation of the domain width, d . Another contribution, $\bar{\mathbf{P}}_{dw}$, is due the DWs motion with the extension of the up- and contraction of the down-oriented domains, while the spontaneous polarization inside domains preserves. As a driving parameter, it is convenient to use the x - y plane coarse-grained electrostatic induction, $\bar{\mathbf{D}} \parallel \mathbf{z}$, which conserves through the paraelectric-ferroelectric interface and therefore is constant across the whole system. Calculation of the linear response of the DW structure, $\bar{\mathbf{P}}_{dw} = \gamma \bar{\mathbf{D}}$ is one of our tasks and will be done below.

The resonance of the system is expected when the frequency of the driving ac field matches the frequency of proper oscillations of DWs, which are described within the harmonic oscillator approximation [17]:

$$\mu \ddot{x}(t) + \eta \dot{x}(t) + kx(t) = 2P_s E_D(t), \quad (2)$$

where x is the coordinate of alternating DW displacements (Fig. 1a), coefficients k , μ and η are calculated per unit of DW area, and $2P_s E_D(t)$ is the pressure of the induction-induced driving field $\bar{\mathbf{E}}_D = (\varepsilon_0 \varepsilon_{\parallel})^{-1} \bar{\mathbf{D}}$, forcing DW to move to flip the surrounding polarization from $-P_s$ to $+P_s$ [22]. *Ab initio* simulations indeed reveal the oscillation dynamics of DWs in the sub-THz range and allow for evaluation of the coefficients of Eq. (2) for thin films of PZT [18]. In what follows we will discuss the physical origin of these parameters and generalize the results of [18] for arbitrary ferroelectric material.

The stiffness constant k is of a purely electrostatic origin and, therefore, is not too sensitive to the profile of DW. Stiffness arises from the restoring force, trying to diminish the depolarizing field, E_{dep} penetrating the film. The latter is caused by depolarizing surface extra-charge $\sigma = \pm \Delta P_s$, induced by the domain spontaneous polarization excess $\Delta P_s = 2 \frac{x}{d} P_s$ and is directed oppositely to ΔP_s : $E_{dep} = -\sigma / \varepsilon_0 = -\frac{x}{d \varepsilon_0} 2P_s$. The depolarizing field and the intrinsic field together form the total field, $E = E_D + E_{dep}$, inside the ferroelectric slab. We evaluate the depolarization energy as the electrostatic energy of the parallel-plate capacitor with the plate area S , carrying the uniformly-distributed charge $Q = \sigma S$, and having the capacity $C = \varepsilon_0 \varepsilon_{\parallel} S / (2a_f)$ as:

$$W = \frac{Q^2}{2C} = \frac{1}{2\varepsilon_0 \varepsilon_{\parallel}} (2a_f) \left(2 \frac{x}{d} P_s \right)^2 S. \quad (3)$$

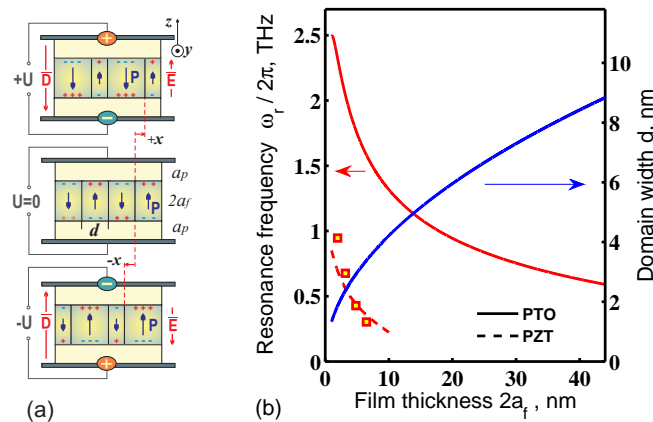


FIG. 1: (a) Oscillation of the periodic domain structure in the sandwiched ferroelectric layer under the applied ac field. (b) Dependence of the domain resonance frequency, ω_r (for PTO and PZT) and of the domain width, d , (for PTO) on the film thickness, $2a_f$. Filled squares show the results of *ab-initio* simulations for PZT [18].

The corresponding energy per unit area of the displaced DW is $w = \frac{d}{S(2a_f)}W$. Relating it with the harmonic oscillator stiffness energy $\frac{1}{2}kx^2$ we express the coefficient k as:

$$k = \frac{4P_s^2}{d\varepsilon_0\varepsilon_{\parallel}}g(z), \quad (4)$$

where the correction factor $g(z)$

$$g(z) = \frac{1}{z} \ln \cosh z \quad (5)$$

with

$$z = z = (\pi\zeta/2) (\varepsilon_{\perp}/\varepsilon_{\parallel})^{1/2} (2a_f/d) \quad (6)$$

was introduced to account for the non-uniform stepwise distribution of depolarization surface charges at termination of alternating domains [17].

The other two coefficients of Eq. (2), effective DW mass, μ , and the viscosity, η , are related with the motion of the material-constituent ions during the variation of the dynamic polarization. Kittel evaluated the DW mass, considering the flipping of polar ions located across propagating DW [23]. Subsequent calculations, however, demonstrated that the effective mass is much larger than that obtained there and depends on the film thickness [17, 18]. This is explained by the larger fraction of participating ions [18] located in the "softer" polarization profile [12, 15] across the entire domain and by the piezoelectric effect of the depolarization field [17]. We adopt here the calculated in [18] effective mass, approximately fitted as $\mu [\text{kg/m}^2] \simeq 1.3\sqrt{2a_f [\text{nm}]} \times 10^{-9}$. Another parameter, viscosity, η , is expressed via DW relaxation time, $\tau \simeq \mu/\eta$, which was also calculated in [18] for PZT films. It was shown, in particular, that τ is naturally related with the soft-mode relaxation of polar ions, τ_i , that is only few times shorter than τ [24].

Equation (2) establishes the response of the DW vibration, $x(t) = x_{\omega}e^{-i\omega t}$, to the periodically applied field, $\mathbf{E}_D(t) = \mathbf{E}_{D\omega}e^{-i\omega t}$. This allows for finding the dynamical relation between Fourier components of the driving induction $\overline{\mathbf{D}}_{\omega} = \varepsilon_0\varepsilon_{\parallel}\mathbf{E}_{D\omega}$ and DW contribution to the ferroelectric layer polarization, $\overline{\mathbf{P}}_{dw\omega}$, that is nothing but the described above polarization excess, $\Delta P_{\omega} = 2P_s \frac{x_{\omega}}{d}$. We obtain the characteristic Lorentzian resonance profile:

$$\overline{\mathbf{P}}_{dw\omega} = \gamma(\omega) \overline{\mathbf{D}}_{\omega}, \quad (7)$$

with

$$\gamma(\omega) = \frac{g^{-1}\omega_0^2}{\omega_0^2 - \omega^2 - i\Gamma\omega}, \quad (8)$$

where

$$\omega_0 = \sqrt{\frac{k}{\mu}} = \left(\frac{4P_s^2 g}{\mu d \varepsilon_0 \varepsilon_{\parallel}} \right)^{1/2} \quad (9)$$

is the natural oscillator frequency. Parameters g and d depend on $2a_f$. The amplitude of the response function, $|\gamma(\omega)|$, reaches its maximum at the resonance frequency $\omega_r^2 = \omega_0^2 - \Gamma^2/2$, see (see Fig. 3a).

Equation (9) enables optimization of the materials parameters and the film thickness to make sure that ω_r falls within the desired THz frequency range. In particular, for the strained films of PTO with the high spontaneous polarization, $P_s \simeq 0.65 \text{ C m}^{-2}$, see Ref. [27], relatively low permittivities $\varepsilon_{\parallel} \simeq 100$, $\varepsilon_{\perp} \simeq 30$, soft mode damping factor $\Gamma \simeq 20 \text{ cm}^{-1}$ (0.6 THz) [26] and μ defined as above, the resonance frequency ω_r decreases and spans the range from 1.5 to 0.75 THz when $2a_f$ increases from 10 to 40 nm.

We find the damping frequency, $\omega_d^2 = \omega_0^2 - \Gamma^2/4$, of the attenuated oscillations of domains in PTO, $x(t) = x_0 e^{-(\Gamma/2)t} \sin \omega_d t$, which is slightly larger than ω_d , see Fig. 3b. Remarkably, our formulas perfectly describe the results of *ab-initio* simulations of DWs oscillations in PZT ultrathin films [18]. The calculated damping frequency, ω_d is shown by the dashed line in the Fig. 3b, the symbols display the results of simulations. Here we used the following parameters for PZT: $P_s \simeq 0.40 \text{ C m}^{-2}$ Ref. [27], $\varepsilon_{\parallel}, \varepsilon_{\perp} \simeq 350$, $\Gamma \simeq 27 \text{ cm}^{-1}$ (0.8 THz) [27] and the same μ . At the same time, the resonance frequency of PZT, ω_r , drops rapidly with the thickness and vanishes above 4 nm.

Having established the dynamical response of the DW-induced polarizations on the applied induction (7), we find the effective dielectric permittivity, $\varepsilon_f(\omega)$. The latter is defined as coefficient of proportionality between induction

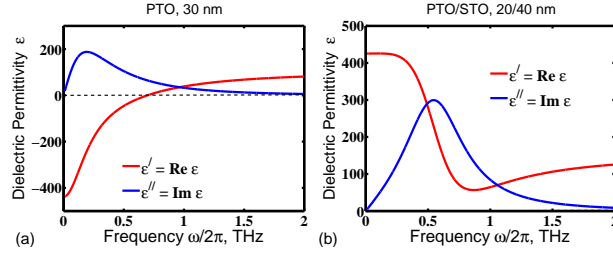


FIG. 2: (a) Dynamic dielectric permittivity of the 30 nm thick film of PTO. (b) The same for either the 20/40 nm sandwich of PTO/STO layers or for the equivalent superlattice.

and the average electric field *inside* the layer, $\bar{\mathbf{D}}_\omega = \epsilon_0 \epsilon_f(\omega) \bar{\mathbf{E}}_\omega$. Taking into account that $\bar{\mathbf{D}}_\omega = \epsilon_0 \epsilon_\parallel \bar{\mathbf{E}}_\omega + \bar{\mathbf{P}}_{dw\omega}$, where the first term is due to intrinsic contribution, $\bar{\mathbf{P}}_i$ we obtain [29]:

$$\epsilon_f(\omega) = \frac{\epsilon_\parallel}{1 - \gamma(\omega)}. \quad (10)$$

The frequency dependence of $\epsilon_f(\omega)$ for 30 nm film of PTO (Fig. 2a) exhibits striking frequency dependence: its real part is negative at low frequencies, and then becomes positive in sub-THz region. This peculiarity already noted in [30] for the static $\epsilon_f(0)$, is explained as a result of the opposite orientation of depolarization field $\bar{\mathbf{E}}_{dep}$ with respect to $\bar{\mathbf{P}} \parallel \bar{\mathbf{D}}$. This field appears to be stronger than the “correctly” oriented driving field $\bar{\mathbf{E}}_D = (\epsilon_0 \epsilon_\parallel)^{-1} \bar{\mathbf{D}}$ and results in orientation of the total field inside ferroelectric slab, $\bar{\mathbf{E}} = \bar{\mathbf{E}}_D + \bar{\mathbf{E}}_{dep}$ against $\bar{\mathbf{D}}$.

This negative- ϵ phenomenon does not lead however to thermodynamic contradiction, since the described domain structure can be observed only when the thickness of the paraelectric buffer, a_p , is larger than the domain width, d , and domain depolarization stray fields do not interact with electrodes [31]. Then, only the effective permittivity, $\bar{\epsilon}$, defined through the total capacity of the system $C = \epsilon_0 \bar{\epsilon} \frac{S}{2a_f + 2a_d}$ makes sense. Under this condition, $\bar{\epsilon}(\omega)$ can be decomposed into two in-series contributions from para- and ferroelectric layers:

$$\frac{1}{\bar{\epsilon}(\omega)} = \frac{\alpha_p}{\epsilon_p} + \frac{\alpha_f}{\epsilon_f(\omega)}, \quad \alpha_{f,p} = \frac{a_{f,p}}{a_f + a_p}. \quad (11)$$

Plot of the $\bar{\epsilon}(\omega)$ dependence for PTO/STO 30/50 nm system with $\epsilon_p = \epsilon_{STO}(1 \text{ THz}) \simeq 200 + 30i$ at 300 K [32] (Fig. 2b) indeed demonstrates the positive $\text{Re } \bar{\epsilon}$ at low ω with the peak at $\simeq 0.5 \text{ THz}$. Making use of expressions (8)-(11) we find:

$$\bar{\epsilon}(\omega) = \bar{\epsilon}(\infty) + \frac{\Delta \bar{\epsilon} \omega_0'^2}{\omega_0'^2 - \omega^2 - i\Gamma\omega} = \frac{\omega_0'^2 - \omega^2 - i\Gamma\omega}{\omega_0'^2 - \omega^2 - i\Gamma\omega} \bar{\epsilon}(\infty), \quad (12)$$

with $\Delta \bar{\epsilon} = \bar{\epsilon}(0) - \bar{\epsilon}(\infty)$, and

$$\begin{aligned} \frac{1}{\bar{\epsilon}(0)} &= \frac{\alpha_p}{\epsilon_p} + \frac{\alpha_f}{\epsilon_\parallel} (1 - \gamma_0), \\ \frac{1}{\bar{\epsilon}(\infty)} &= \frac{\alpha_p}{\epsilon_p} + \frac{\alpha_f}{\epsilon_\parallel}. \end{aligned} \quad (13)$$

The effective oscillator frequency: $\omega_0' < \omega_0$ satisfies the Lyddane-Sachs-Teller relation

$$\frac{\omega_0'^2}{\omega_0^2} = \frac{\bar{\epsilon}(\infty)}{\bar{\epsilon}(0)} = 1 - \alpha_f \frac{\epsilon(\infty)}{\epsilon_\parallel} \gamma_0 \quad (14)$$

and can be tuned by the geometrical parameters α_p, α_f .

The classical impedance-spectroscopy setup of Fig. 1a, used so far for calculations of $\epsilon_f(\omega)$, is not suitable for the experimental study in sub- and low-THz region, where the far-infrared and THz-optics can be explored [1, 2, 19]. The low-THz resonance in ultrathin films of $2 \div 10 \text{ nm}$, where the resonance frequency is only slightly smaller than the frequency of the soft mode $E(TO_1)$, can be also captured by Raman spectroscopy. In the reflection-absorption geometry shown in Fig. 3, the incident p - (in-plane) polarized THz beam is reflected from the STO substrate through

the PTO film and the intensity of the outgoing radiation is measured. The dynamics of domain structure can be detected by its interaction with the \mathbf{z} -component of the field, \mathbf{E} , when the reflected beam passes twice through the PTO layer.

The THz reflectance of the thin film with $2a_f < \lambda = 2\pi c/\omega$, where the wavelength $\lambda \simeq 1 \div 0.15$ mm for $0.3 \div 2$ THz radiation, deposited on the thick substrate with $a_p > \lambda$, is calculated from $\varepsilon_f(\omega)$ as [19]:

$$R^{(p)} \approx R_0^{(p)} \left| 1 - \frac{2a_f}{\lambda} \frac{8\pi \cos \theta \sin^2 \theta}{\left(\cos \theta - \varepsilon_p^{-1/2} \sin \theta \right)^2} \text{Im} \frac{1}{\varepsilon_f(\omega)} \right|, \quad (15)$$

where θ is the incidence angle and

$$R_0^{(p)} = \left| \frac{\varepsilon_p \cos \theta - \sqrt{\varepsilon_p - \sin^2 \theta}}{\varepsilon_p \cos \theta + \sqrt{\varepsilon_p - \sin^2 \theta}} \right|^2 \quad (16)$$

is the reflectance of the substrate alone.

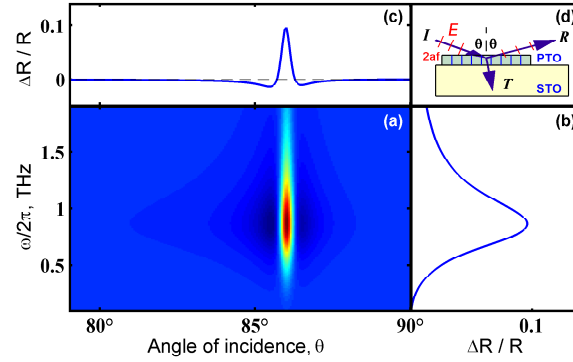


FIG. 3: Calculated reflectivity, $\Delta R/R$, for the p -polarized beam of sub-THz radiation of $2a_f \simeq 30$ nm PTO film with domains, deposited on the thick STO substrate, as function of the incidence angle, θ , and the beam frequency, ω . The resonance enhancement is observed at $\omega_r/2\pi \simeq 0.75$ THz and at the Brewster incidence angle $\theta_B \simeq 86^\circ$. (a) Two-dimensional color map of $\Delta R/R$ dependence of θ and ω . (b) Cross-cut of $\Delta R/R$ at $\theta = \theta_B$. (c) Cross-cut of $\Delta R/R$ at $\omega = \omega_r$. (d) The geometry of the experiment.

To identify the contribution of the domain structure we consider the thin 30 nm film of PTO deposited on thick, ~ 0.5 mm substrate of STO. The frequency and the incidence angle dependencies of reflectivity, $\Delta R/R = (R_0^{(p)} - R^{(p)})/R_0^{(p)}$, [19] is shown in Fig. 3. The domain-provided enhancement of reflectivity of about $\Delta R/R \simeq 0.1$, is observed at $\omega/2\pi \simeq 0.75$ THz at the Brewster incidence angle $\theta_B \approx \arctan(\text{Re} \varepsilon_p)^{1/2} \simeq 86^\circ$, when the reflectance of p -wave from the substrate alone, $R_0^{(p)}$, is as small as the imaginary part of ε_{STO} . The incident beam almost tangential to the surface can be realized by placing the high-refractive Otto prism at the top of the sample [19]. Importantly, even better result can be achieved by using the PTO/STO superlattice instead of PTO monolayer. The reflectivity, $\Delta R/R$, will increase proportionally to the number of PTO layers and can become giant at $\theta = \theta_B$.

To conclude, we calculated the dynamic dielectric permittivity $\varepsilon_f(\omega)$ of the regular structure of 180° domains in strained ferroelectric films and superlattices. We demonstrated that the collective vibrational mode with the resonance frequency $\omega_r/2\pi \simeq 0.3 \div 3$ THz can be detected in PTO/STO systems by means of the reflection-absorption spectroscopy. This unique property makes ferroelectric films a promising candidate for compact and tunable devices working in the sub- and low THz range.

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Supplementary Material: Static Permittivity of Ferroelectric Film with 180° Domains

Static permittivity of the ferroelectric film with domains, sandwiched between two paraelectric layers was calculated by A. Kopal, P. Mokřý, J. Fousek and T. Bahník [29] as:

$$\varepsilon_{\text{eff}} = \varepsilon_z^{(2)} \frac{D}{d'} \left(1 + B \frac{\varepsilon_z^{(2)}}{\varepsilon_z^{(1)}} \right)^{-1} + \left[\left(1 + B \frac{\varepsilon_z^{(2)}}{\varepsilon_z^{(1)}} \right) \left[\frac{B}{(1+B)\varepsilon_z^{(1)}} - \frac{2 \ln 2}{R_{\text{eq}}^0 (1+B)(g^{(1)} + g^{(2)})} \left(1 + B \frac{\varepsilon_z^{(2)}}{\varepsilon_z^{(1)}} \right) \right] \right]^{-1}, \quad (17)$$

In our notations,

$$\begin{aligned} \varepsilon_{\text{eff}} = \varepsilon_{\text{tot}}(0), \quad \varepsilon_z^{(1)} = \varepsilon_p, \quad \varepsilon_z^{(2)} = \varepsilon_{\parallel}, \quad g^{(1)} + g^{(2)} = \varepsilon_p + (\varepsilon_{\parallel} \varepsilon_{\perp})^{1/2} = \varsigma (\varepsilon_{\parallel} \varepsilon_{\perp})^{1/2}, \\ \frac{d'}{D} = \frac{2a_f}{2a_p + 2a_f} = \alpha_f, \quad B = \frac{2a_p}{2a_f} = \frac{\alpha_p}{\alpha_f}, \quad R_{\text{eq}}^0 = \frac{\pi}{2} \frac{2a_f}{d}, \end{aligned} \quad (18)$$

Eq. (??) can be written as:

$$\varepsilon_{\text{tot}}(0) = \left(1 + \frac{2a_p}{2a_f} \frac{\varepsilon_{\parallel}}{\varepsilon_p} \right)^{-1} \left(\frac{\varepsilon_{\parallel}}{\alpha_f} + \left[\frac{\alpha_p}{\varepsilon_p} - \frac{\alpha_f}{\varepsilon_{\parallel}} \frac{4 \ln 2}{\pi \varsigma} \left(\frac{\varepsilon_{\parallel}}{\varepsilon_{\perp}} \right)^{1/2} \frac{d}{2a_f} \left(1 + \frac{2a_p}{2a_f} \frac{\varepsilon_{\parallel}}{\varepsilon_p} \right) \right]^{-1} \right). \quad (19)$$

The inverse permittivity, after some algebra, can be decomposed onto two in-series contributions from paraelectric and ferroelectric layers,

$$\frac{1}{\varepsilon_{\text{tot}}(0)} = \frac{\alpha_p}{\varepsilon_p} + \frac{\alpha_f}{\varepsilon_f(0)} \quad (20)$$

where the permittivity of ferroelectric layer,

$$\varepsilon_f(0) = \varepsilon_{\parallel} - \frac{\pi \varsigma}{4 \ln 2} \left(\frac{\varepsilon_{\perp}}{\varepsilon_{\parallel}} \right)^{1/2} \frac{2a_f}{d} \varepsilon_{\parallel}, \quad (21)$$

has the positive (intrinsic) contribution and the negative (DW-provided) contribution.